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Report N00014-79-C-0488

COHERENT TRANSIENT EFFECT STUDIES OF RYDBERG ATOMS

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25 March 1981

Interim Report for Period 1 March 1980 - 1 March 1981

Unlimited Distribution

Prepared for
OFFICE OF NAVAL RESEARCH
Department of the Navy
Arlington, VA 22217



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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER N00014-79-C-0488-2	2. GOVT ACCESSION NO. AD 140 97588	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) "Coherent Transient Effect Studies of Rydberg Atoms"	5. TYPE OF REPORT & PERIOD COVERED 12/12	
6. AUTHOR(s) Richard L. Shoemaker William H. Wing	7. CONTRACT OR GRANT NUMBER(s) N00014-79-C-0488	
8. PERFORMING ORGANIZATION NAME AND ADDRESS Physics Dept./Optical Sciences Center University of Arizona Tucson, AZ 85721	9. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS RR 011 03 01 NR 393-011	
10. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research Dept. of the Navy Arlington, VA 22217	11. REPORT DATE 25 March 1981	
12. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	13. NUMBER OF PAGES 5 pages	
14. DISTRIBUTION STATEMENT (of this Report) Unlimited distribution	15. SECURITY CLASS. OF THIS REPORT Unclassified	
16. DECLASSIFICATION/DEGRADING SCHEDULE	17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)	
18. SUPPLEMENTARY NOTES A	19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Coherent transient effects, Rydberg atoms, optical coherent transients, highly excited atoms.	
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) New techniques for making time-resolved studies of atoms in highly excited (Rydberg) states are being developed. In particular, the absorption or emission of radiation by Rydberg-state atoms is monitored on a nanosecond time scale during and after irradiation by various CO ₂ laser pulse sequences. The signals thus produced (known as coherent transient effects), will be studied in order to provide both information about collisional relaxation processes in Rydberg atoms and other spectroscopic data.		

Contract Description

New techniques for making time-resolved studies of atoms in highly excited (Rydberg) states are being developed. In particular, the absorption or emission of radiation by Rydberg-state atoms is monitored on a nanosecond time scale during and after irradiation by various CO₂ laser pulse sequences. The signals thus produced (known as coherent transient effects), will be studied in order to provide both information about collisional relaxation processes in Rydberg atoms and other spectroscopic data.

Scientific Problem

A detailed knowledge of collisional relaxation and energy transfer in atomic Rydberg states is important in a variety of areas, ranging from atomic plasma behavior to the construction of atomic Rydberg lasers and the use of Rydberg atoms as single quantum detectors of microwave radiation. Although there is currently a great deal of activity in the field of Rydberg atoms, almost all of our knowledge about their collisional relaxation comes from line-broadening studies rather than from direct time-resolved experiments. We are attempting to do direct experiments of this sort. We anticipate that these studies will yield much more detailed information about collisional cross sections for specific relaxation channels as well as other useful spectroscopic data such as precise transition frequencies and absolute values for Rydberg transition dipole matrix elements.

Scientific and Technical Approach

In order to study Rydberg states with coherent transient effects, the following approach is being employed: A pair of synchronously-pumped pulsed dye lasers excite ground-state atoms to a Rydberg level via resonant two-step excitation. At the same time, the atoms are illuminated with a cw CO₂ laser which can be brought into resonance with the Rydberg transition of interest by an electric field applied to the sample. Immediately following the dye laser excitation, transient effects are produced in the sample by pulsing the electric field in order to shift the Rydberg atoms in and out of resonance with the CO₂ laser. By monitoring the transmitted CO₂ laser beam, effects such as photon echoes, optical nutation, adiabatic rapid passage, and others can be observed, depending on the pulse sequence one applies. We hope to be able to measure absolute transition dipole matrix elements, accurate Stark shift coefficients, Rydberg transition frequencies, and cross-sections for specific collisional relaxation processes. In addition, we hope to measure collisional velocity redistribution effects and the velocity dependence of collisional relaxation rates.

Progress

The entire apparatus needed to do the Rydberg coherent transient experiments along with a variety of diagnostic tools has been constructed. The performance of all the component pieces has also been tested both individually and as part of an integrated system. Performance of the major components is as follows: The two dye lasers provide 1 to 2 kW peak power with a bandwidth of ~ 8 GHz in a 5 nsec. pulse. The CO₂ laser provides 1 to 2 W of single mode TEM₀₀ power with short term jitter of less than 50kHz and long term stability of ~ 2 MHz/hr. The sample cell maintains the alkali metal vapor (currently

sodium) at a known, controllable pressure, transmits both the infrared and visible laser beams, and contains a pair of precision Stark plates to which both a dc bias and pulsed electric fields can be applied. The cell also incorporates an ultraviolet fluorescence detection system for monitoring Rydberg populations and a Faraday cup ion detection system for detecting Rydbergs via field or collisional ionization. Since there are a large number of possible Rydberg transitions and CO₂ laser lines, a number of computer programs have been written and used to guide us in determining which transitions to investigate. Finally, a microcomputer system interfaced to a transient recorder has been built for data acquisition and signal averaging.

A number of experiments have been performed to verify that everything is working properly. With sodium in the cell, strong fluorescence signals from the 4P → 3S transition at 3303Å (the 4P state is populated in cascade from higher levels) are observed when the first dye laser is tuned to the 3S_{1/2} → 3P_{3/2} transition and the second to a 3P_{1/2} → nD or nS transition. The second transition can also be shifted out of resonance by applying a field to the Stark plates, thus verifying that they work properly. When both dye lasers are resonant, we also observe some interesting and unexpected signals from the Faraday cup ion collection system. These signals, which depend strongly on L and only weakly on n, appear to arise from collisional ionization of the Rydbergs. We hope to study this effect further in the near future.

A number of attempts have been made to observe the coherent transient signals, but we have not succeeded so far. Since all the available evidence we have indicates that the apparatus meets the requirements needed to see such signals, we presently believe that we simply have a search problem. Our calculations, based on standard tabulated transition data, are of marginal accuracy for determining CO₂ laser - atomic transition coincidences. We are currently working to improve these calculations, as well as continuing our experimental searches.

Publications: None as yet

Unspent Funds: We do not anticipate any funds to remain unspent at the end of the current contract period.

Graduate Students: Carl Gaebe

No degrees awarded as yet.

A. Richard L. ShoemakerCurrent Support

NSF grant CHE-7824085, "Molecular relaxation and coherent transient effects"; R.L. Shoemaker, principal investigator; 6/1/80 - 5/31/82, \$49,539 per year, 20% effort.

ONR contract N00014-79-C-0488, "Coherent transient effect studies of Rydberg atoms"; W.H. Wing and R.L. Shoemaker co-principal investigators; 6/1/79 - 5/31/81, \$62,770 per year, 20% effort.

NIH contract 1-R01CA24466-01, "Ultrafast scanner microscope in laboratory automation"; R.V. Shack, principal investigator; 5/1/78 - 4/30/81, \$174,500 per year, 25% effort.

NSF grant PHY-7915302, "Modern spectroscopy and collision studies of fundamental atomic and molecular systems"; W.H. Wing, principal investigator; 11/18/80 - 11/17/81, \$150,000, 1% effort (Dr. Shoemaker is included in this grant so that a faculty member is available to coordinate paperwork and provide signatures during periods when Dr. Wing is out of town). .

B. William H. WingCurrent Support

NSF grant PHY79-15302, "Modern Spectroscopy and Collision Studies of Fundamental Atomic and Molecular Systems," W. H. Wing and R. L. Shoemaker, co-principal investigators; 11/1/80 - 10/31/81, \$150,000, 60% effort.

PRF (American Chemical Society) 12206, "Laser Spectroscopy and Collision Studies of Molecular Ions," W. H. Wing, principal investigator, 3/1/80 - 8/31/82, \$30,000, 5% effort.

ONR contract N00014-79-C-0488, "Coherent transient effect studies of Rydberg atoms"; W. H. Wing and R. L. Shoemaker, co-principal investigators; 6/1/79 - 5/31/81, \$62,770 per year, 25% effort.

NBS NB80NADA1050, "Electrostatic Trapping of Neutral Atomic Particles," W. H. Wing, principal investigator; 9/28/80 - 9/27/81, \$25,000, 10% effort.

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